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The reliability study on the Cu/CHA NH_3-SCR catalysts: SO_3 and Na ions poisoning

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The deactivation mechanism of Cu/CHA ammonia selective catalytic reduction catalysts by SO_3 poisoning has been systematically investigated using a range of analytical techniques. In order to study the influence of SO_3 poisoning on active Cu^{2+} ions and the zeolite framework, different sulfate samples were prepared with different contents of SO_3 (0-20%) in SO_x under same poisoning condition. The results reveal the NO conversion of samples poisoned by SO_3 decreased more than that poisoned by SO_2 when temperature ranged between 100° and 600°. The TPR and EPR results demonstrate that SO_3 poisoning does a significant influence on the amount of active Cu^{2+} ions than SO_2 does. The kinetic results illustrate the SO_3 poisoning has no impact on the apparent activation energy (Ea) of NH3-SCR reaction over Cu/CHA catalysts. The reason of NH_3-SCR activity declining is the reduction of the number of isolated Cu^{2+} ions among the kinetic temperature regions. The *ex-situ* DRIFTs and BET results expose that the SO_3 poisoning could decrease the crystallization by damaging Si-OH-Al structure. The NH_3-SCR activity at high temperature decline because of the NH_3 migration difficulty resulted by structure damaging. Cu/CHA catalysts have been found to be affected by alkali and alkali earth ions; however, the poisoning mechanism is still unclear. In order to investigate Na poisoning effects and its mechanism on Cu/SAPO-34 and Cu/SSZ-13, five samples with different Na contents were synthesized. The Na effects on the structure, Cu species, and NH_3-SCR reaction over Cu/CHA were characterized through XRD, BET, NH_3-TPD, ex-DRIFTS, H_2-TPR, EPR, activity tests and kinetic experiments, and CO_2-DRIFTS were used to probe the types of Na species. The results indicate that the introduced Na^+ exchanged with H^+ and Cu^{2+}, and it mainly substituted H^+ from Si-OH-Al, then H^+ from surface OH, finally isolated Cu^{2+}. The exchanged H^+ led to the structure damaging of Cu/CHA by dealumination, and the exchanged Cu^{2+} aggregated and formed CuOx species. The NH_3-SCR activity decreased with Na contents, and the loss of isolated Cu^{2+} and CHA structure was responsible for the performance deactivation.

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